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PICATINNY ARSENAL TECHNICAL REPORT 3112
STEADY-STATE CONDITIONS
IN AN
EXPLOSIVE WHICH IS SUBJECTED EXTERNALLY
TO
ELEVATED TEMPERATURES

FRED P. STEIN

MARCH 1965

PICATINNY ARSENAL
DOVER, NEW JERSEY

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(6) STEADY-STATE CONDITIONS IN AN EXPLOSIVE
WHICH IS SUBJECTED EXTERNALLY
TO ELEVATED TEMPERATURES ,

(10) by

Fred P. Stein

(11) March 1965,

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PREFACE

This report has two distinct sections, a longer-than-usual abstract which summarizes the author's findings and feelings on the steady-state problem and a somewhat detailed treatment of the same matter. The former is commended to all readers, and the latter to those inspired by the abstract and to anyone who is going to take up this type of study. For such an individual, the latter section could save months of time.

This report contains, in far greater detail than could be found in the literature, a critical appraisal of the tools available for handling the steady-state problem, as to (a) usefulness, (b) validity of assumptions contained in the procedures, and (c) problems left in an unsatisfactory state requiring a research effort.

A second report on the subject of time to explosion after receipt of a thermal stimulus is in preparation.

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ABSTRACT

The question "Will an explosive explode when subjected to a given elevated temperature?" can be attacked by studying the heat transfer in the explosive if the question is rephrased to read "Is it possible to realize steady-state temperature gradients in an explosive which is subjected to a given elevated temperature?" If the answer is no, then the resulting "runaway" temperature profiles signal that an explosion will occur.

The steady-state heat-transfer equation containing a term for heat generation by chemical reaction at all points was studied from a number of points of view. The heat-generation term was governed by a zero-order rate-of-reaction equation. With this type of kinetic expression, the explosion condition can be obtained rigorously from the mathematics without defining what kind of temperature rise represents an explosion. The temperature variance of the zero-order rate equation was described by the Arrhenius expression, which in turn was approximated by the so-called "exponential approximation." It was shown that this approximation is satisfactory for the steady-state problem and that it no longer need be questioned in connection with this problem. The physical parameters were all considered not to be functions of temperature. Again, this approximation is adequate for steady-state problems where the temperature differences involved are not large. Where a steady state was calculated to exist no temperature differences in the explosive in excess of 10°C were observed.

Of primary concern in the steady-state problem is the minimum temperature of explosion. If the environment of the explosive is below the minimum temperature of explosion, steady-state temperature gradients can exist in the explosive; if the environmental temperature is above, then an explosion occurs. It is to be emphasized that the minimum temperature of explosion is not a property of the explosive material itself, for it is quite dependent on the environment, mass, and configuration of the explosive charge. The following table shows minimum temperatures of explosion for lead azide which were calculated in the course of this work. The values range from 149°C to 269°C . It was presumed that the activation energy ($E = 36,300$ cal/g mole), frequency factor ($Z = 10^{12.0}$ sec $^{-1}$), and heat of explosion ($Q = 399$ cal/g) are intrinsic properties of lead azide with the correct numerical values

as stated. The variations among the values in the table resulted from variations in the thermal conductivity of the powder bed of lead azide (λ), the bulk density of the powder bed (ρ), and the thickness of material (a), with environmental conditions and mass of lead azide considered. The numerical values given to these properties are thought to be realistic. The thermal conductivity of beds of very fine powder is very difficult to determine. The great range of values appearing in the table reflects attempts to estimate the thermal conductivity of the lead azide powder bed with either helium at atmospheric pressure or air at 0.4-micron pressure in the interstices. The crystal density of lead azide is 4.705 g/cm^3 ; however, photographs of lead azide involved in ignition experiments revealed its bulk density to be 0.54 g/cm^3 in that situation. Hence, the bulk densities used are in the correct range. The thickness can, of course, be as desired.

TABLE 1

Minimum Temperature of Explosion of Lead Azide

<u>Tm ($^{\circ}\text{C}$)</u>	<u>λ (cal/cm sec $^{\circ}\text{K}$)</u>	<u>ρ (g/cm³)</u>	<u>a (cm)</u>
149*	2.51×10^{-8}	0.54	0.078
173	1.55×10^{-5}	0.88	0.509
184*	2.51×10^{-8}	0.54	0.078
187	1.55×10^{-4}	2.64	0.509
192	1.55×10^{-4}	1.76	0.509
201	1.55×10^{-4}	0.88	0.509
233	1.55×10^{-3}	0.88	0.509
263*	1.55×10^{-4}	0.54	0.078
264*	1.55×10^{-4}	0.54	0.078
269	1.55×10^{-3}	0.88	0.051

* See text for explanation of difference.

It is interesting to compare the first and third Tm's, 149°C and 184°C , in Table 1. Since all parameters are identical, the results would be expected to be the same. However, different boundary conditions were used in the two cases. The first was calculated under the conditions that the lower surface of an infinite slab 0.078 cm thick was held at a constant temperature

while the upper surface was allowed to communicate heat with an environment at 50°C governed by a heat-transfer coefficient of $2 \times 10^{-4} \text{ cal/cm}^2 \text{ sec}^\circ\text{C}$. It should also be pointed out that for these parameters an increase in the heat-transfer coefficient to infinite value, which is tantamount to maintaining the upper surface at 50°C, would not increase the minimum temperature of explosion above 184°C.

The effect of the boundary conditions is also available for the T_m 's of 263°C and 264°C, the former being for the perfect-insulation boundary condition. Here, with the greater thermal conductivity, the heat transfer at the upper surface hardly affects the minimum temperature of explosion.

The solutions to the steady-state problem which appear in the literature have been given a critical review. It was concluded that the solutions are adequate and usable. Although it is possible to construct boundary conditions for which solutions are not available, there currently does not appear to be any set of conditions for which a solution should be obtained forthwith.

During this study of the steady-state problem, a few things were developed which are improvements over the current literature; however, since they concern steady-state profiles, they are secondary in importance to the main objective of defining critical conditions for explosion. The equation shown in this report for obtaining a steady-state temperature profile in the case of symmetrical heating (lower face of slab at constant temperature, upper face perfectly insulated) is the best solution available for this problem; for the case of unsymmetrical heating (lower face at constant temperature, upper face communicating with the environment), the equations developed in this report are more easily and more rapidly handled than those appearing in the literature.

Daniel S. Ling, Jr., of P.E.C. Corporation is currently developing a theory of initiation of explosives using a statistical approach. The first adequate summary of his progress to date has recently been published in Proceedings of the Eleventh Basic Research Group Contractors' Conference and Symposium, U.S. Army, ERDL, pp, 225-271, 1962 and in the final report submitted by P.E.C. Corporation on Contract DA-44-009-ENG-4774. Those papers have not been studied in minute detail, but it is recommended

that the Explosives Research Section retain scientific cognizance of the developments. Currently, numerical calculations are difficult or impossible with merely the referenced article. Calculations were attempted for lead azide, but the parameters were far out of range of those listed by Ling. The illustrative computations in his article merely show that, at the current stage of development, correct "ball park" answers result for a "typical" explosive.

INTRODUCTION

In the study of steady-state heat conduction in explosives, the limiting conditions under which a steady state can exist are of primary importance. That is to say, if the temperature of the environment of a specific explosive in a specified condition is continuously increased in a stepwise fashion, at each step a steady-state temperature distribution will be set up in the explosive; however, these temperature increases cannot continue indefinitely, for at some temperature a steady-state will no longer be possible, and an explosion will result. It is this limiting temperature for the steady state, called the minimum temperature of explosion, which is of prime interest. Of course, the temperature could be considered constant, and some other condition could be varied until an explosion resulted.

Nearly all of the available literature pertinent to this subject has been covered; however, not all of the literature which was inspected is summarized in this report. The following works are of particular interest: (1) Finklestein and Gamow (NAVORD Report 90 - 46, 20 Apr 1947) is fine for original guidance, (2) Frank-Kamenetsky (Zhurnal Fizicheskoy Khimii, XXXII, 1182, 1958) is credited by most authors with originating the critical-conditions-for-explosion concept, (3) Chambre (J Chem Phys, 20, 1795, 1952) gives an excellent account of solutions for critical conditions, (4) a series of articles by Thomas, and sometimes co-workers (Trans Far Soc, 2007, 1961) gives the most complete and most realistic treatment, and (5) Gray and Harper (Trans Far Soc, 55, 581, 1959) give an interesting account.

The numerical calculations which are considered were made for slabs of RDX 2.5 cm thick and infinite in all other dimensions and for slabs of lead azide in a variety of dimensions. The physical and chemical parameters used for RDX are listed by Zinn and Mader in J Appl Phys, 31, 323, 1960. The parameters for lead azide are given near the mention of the calculations. Credit goes to Peter McIntyre, Cooperative Student from the University of Detroit, for plowing through the detailed numerical work and plotting the graphs. His efforts contributed materially to the writing of this summary.

THE PROBLEM

The time-varying problem of heat conduction in an explosive is expressed as follows:

$$\rho Q Z e^{-E/RT} = -\lambda \nabla^2 T + \rho C \frac{\partial T}{\partial t} \quad (1)$$

The steady-state condition imposes the restriction of $\frac{dT}{dt} = 0$ in Equation 1. Thus, the equation of interest for the steady state becomes

$$\rho Q Z e^{-E/RT} = -\lambda \nabla^2 T \quad (2)$$

Two sets of boundary conditions were considered in detail. The conditions which are easiest to handle are:

$$T(\pm a) = T_{HP} \quad (3)$$

$$\left(\frac{dT}{dx} \right)_{x=0} = 0 \quad (4)$$

These conditions require that the temperature profile in an infinite slab be symmetrical about the center plane which is taken as the $x = 0$ position.

$$T(0) = T_{HP} \quad (5)$$

$$-\lambda \left(\frac{dT}{dx} \right)_{x=2a} = h (T_{2a} - T_{\infty}) \quad (6)$$

Equations 5 and 6 lead to unsymmetrical temperature profiles which make the problem more difficult to handle; however, these conditions are quite realistic for experimental conditions in which the upper (here $x = 2a$) face of the "slab" is exchanging heat with a gaseous or "vacuum" medium and the lower face (here $x = 0$) is in contact with a hot surface.

Another set of boundary conditions was considered briefly. A combination of the conditions expressed in Equations 4 and 6 is a consistent set, if Equation 6 is required to hold for both faces of the slab at $x = \pm a$, the center plane being the $x = 0$ position. The difficulty with all boundary conditions involving a heat-transfer coefficient h is getting a numerical value for h in which one has confidence.

Equation 2 was solved for one-dimensional heat flow in an infinite slab, which makes the Laplacian operator, $\nabla^2 T$, equal to $\frac{d^2 T}{dx^2}$, with the assumptions that reactant consumption is

negligible and that the physical properties of the explosive do not vary with temperature.

APPROXIMATIONS FOR THE RATE OF REACTION TERM

If Equation 2 for one-dimensional heat flow in a slab is changed to dimensionless variables, Equation 7 results.

$$\frac{d^2\theta}{d\xi^2} = -e^{-\frac{1}{\theta}} \quad (7)$$

where

$$\theta = \frac{RT}{E} \quad \text{and} \quad \xi = \left[\frac{R\rho QZ}{E\lambda} \right]^{1/2}$$

The solution to Equation 7 in closed form has not appeared in the literature. Furthermore, it is believed that Equation 7 cannot be solved analytically in closed form.

As a result of this inability to handle Equation 7, at least two approximations of the exponential term in the Arrhenius expression have been used. The exponential approximation has been most widely used and is investigated thoroughly in this report; the quadratic approximation has been used less frequently and receives only passing attention here.

Although very few authors so state and some even attach a degree of mystery to it, the exponential approximation is actually a Taylor-series expansion for $1/T$ about the point $1/T_{HP}$, neglecting all but the first two terms. Thus,

$$\frac{1}{T} \approx \frac{1}{T_{HP}} - \frac{1}{T_{HP}^2} (T - T_{HP}) \quad \text{which leads to the approximation}$$

$$e^{-E/RT} \approx \left(e^{-E/RT_{HP}} \right) \left(e^{\frac{E(T - T_{HP})}{RT_{HP}^2}} \right)$$

It is reasonable to ask how well the approximation describes the actual functions. Mathematical manipulation of the approximation for $1/T$ results in

$$1/T \approx \frac{2T_{HP} - T}{T_{HP}^2}$$

Figure 1 shows this approximation for hot-plate temperatures of 380°C and 340°C. It can be seen on the graph that the true values for $1/T$ form a hyperbola which is approximated by two straight lines, one for $T_{HP} = 380^\circ\text{C}$ and one for $T_{HP} = 340^\circ\text{C}$. Each straight line gives the exact value at its hot-plate temperature. It should be noted that the difference between the approximation and the function is 3.0% at 100°C below T_{HP} of 380°C and is 5.3% at 150°C below the hot-plate temperature. Similarly, it can be seen that for a hot-plate temperature of 340°C, the difference is 7.1% at 160°C above the hot-plate temperature.

It can be concluded from Figure 1 that $1/T$ is always greater than the approximation. Therefore, the "true" reaction rate, which is proportional to $e^{-1/T}$, will always be less than the rate given by the approximation. Thus, if the approximated rate is too great, the approximated minimum temperature of explosion will be lower than the true minimum temperature of explosion. Computed minimum temperatures of explosion always seem to be lower than experimentally observed ("true") minimum temperatures of explosion; however, the differences are much too large to be ascribed to this approximation alone.

It was interesting to compare absolute values of $1/T$ with the approximation; however, it may be clearer to compare the ratio of the approximation with $1/T$. Figure 2 shows this ratio as a function of temperature. Obviously, the further one is from the hot-plate temperature, the poorer is the approximation. On this graph, the ordinate can be read directly as a fraction of the true value. It can be seen that the approximation is within 99% of the true value up to approximately 60°C from the hot-plate temperature and that the distribution is essentially symmetrical about the hot-plate temperature.

Since the term $e^{-E/RT}$, and not $1/T$ appears in Equation 2, a better comparison can be made between $e^{-E/RT}$ and its approximation $\left(e^{-E/RT_{HP}} \right) \left(\frac{E(T - T_{HP})}{RT_{HP}^2} \right)$. Figure 3 shows this comparison for an activation energy of 65,000 cal/g mole at hot-plate temperatures of 340°C and 380°C. As has been indicated earlier in this report, the approximated rate is always greater than the "true" rate and its distribution is approximately symmetrical about the hot-plate temperature. It should be noted

that the percentage error for a given temperature interval away from the hot-plate temperature is much more severe than for the comparison of the exponent made in Figures 1 and 2. The "true" rate will be approximated within 5% at temperature differences of less than 15°C from the hot-plate temperature. If the temperature difference is increased to 25°C, the approximation will give the "true" rate within 10%.

Gray and Harper, in *Trans Far Soc* 55, 581 (1959), present an approximation for $e^{-E/RT}$ which differs from the previous one. It is referred to as the "quadratic approximation": $e^{-E/RT} \approx$

$$e^{-E/RT_{HP}} \left\{ 1 + (e - 2) \phi + \phi^2 \right\} \quad \text{where} \quad \phi = \frac{E}{RT_{HP}^2} (T - T_{HP}).$$

Figure 3 shows this approximation for an activation energy of 65,000 cal/g mole (which, when this computation was made, was believed to be the best number for lead azide) at a hot-plate temperature of 380°C. The approximation is only reasonable for temperatures above the hot-plate temperature, but this limitation is not restrictive for steady-state calculations because all temperatures in the explosive are greater than the hot-plate temperature at steady state. It appears from Figure 3 that the quadratic approximation is not quite as good as the exponential approximation for temperatures quite close to that of the hot plate. For temperatures ranging from about 10°C to 25°C above the hot-plate temperature the quadratic approximation is better. At temperatures more than 25°C above the hot-plate temperature, the quadratic approximation is very bad.

SOLUTION TO STEADY-STATE PROBLEM USING APPROXIMATIONS FOR THE RATE-OF-REACTION TERM

Since it is apparently not possible to solve Equation 2 as it stands, the exponential approximation to the term $e^{-E/RT}$ was made. This equation was then transformed into one with dimensionless variables. The result is Equation 8.

$$\frac{d^2\phi}{dy^2} + k \frac{d\phi}{dy} = -\delta e^{\phi} \quad (8)$$

where $k = 0$ for an infinite slab, $k = 1$ for an infinite cylinder and $k = 2$ for a sphere. Since only infinite-slab geometry was considered, $k = 0$ in all of the work described in this report. If k equals the integers one and two, the mathematical solutions become progressively more complex.

Equation 8 has been solved for infinite-slab geometry using the boundary conditions shown in Equations 3 and 4 in dimensionless form. The mechanics of the solution are available from the author on request. The solution is:

$$(y-1) = \sqrt{\frac{2e^{-\phi_0}}{\delta}} \left[\tanh^{-1} \sqrt{1 - e^{-\phi_0}} - \tanh^{-1} \sqrt{1 - e^{\phi - \phi_0}} \right] \quad (9)$$

Equation 9 is better than any solution currently available in the literature for getting a temperature profile in an explosive at steady state for the particular boundary conditions used. In fact, of all the other authors, only Finklestein and Gamow (NAVORD Report 90-46, 20 April 1947) even list the solution for getting the steady-state profile. The procedure for getting a temperature profile from Equation 9 involves two steps. First, the temperature at the center plane is found by setting $y = 0$, for which ϕ becomes ϕ_0 . The resulting equation is solved for ϕ_0 , the dimensionless center-plane temperature. This number for ϕ_0 is used in Equation 9, which now has only y and ϕ as unknowns; that is to say, one can pick a value of ϕ and calculate the corresponding y to get a temperature-distance curve.

Chambre (in J Chem Phys 20, 1795, 1952) showed solutions to the same problem and same boundary conditions for infinite-cylinder geometry and spherical geometry. His article is beautifully written and, although Frank-Kamenetsky is usually credited with being first with a solution of the problem of infinite-slab geometry, it is Chambre's who should be credited for a fine, clear job with the infinite cylinder and sphere in contrast to Frank-Kamenetsky's crude, sketchy articles. Chambre's solution for the infinite cylinder is shown in Equation 10.

$$\phi = \frac{\ln \frac{8B}{\delta}}{(B\bar{Z}^2 + 1)^2} \quad (10)$$

where $\frac{(8B/\delta)}{(B+1)^2} = 1$ is used to get B.

The solution for the sphere ($k = 2$ in Equation 8) is considerably more complicated. Chambre gives the solution in his article in the form of a tabulated function.

Thomas (in Trans Far Soc 54, 60, 1958) has solved Equation 8 with boundary condition 6 in dimensionless form applying at both faces of the infinite slab and on the surface of the infinite cylinder, which of course requires boundary condition 4 in dimensionless form to apply at the centers.

Thomas and Bowes solved Equation 8 with dimensionless boundary conditions shown in Equation 5 and 6 for an infinite slab, the most realistic case for the experimental setup used by the Explosives Research Section at Picatinny. This solution is discussed in more detail in the section of this report entitled "Review of Thomas and Bowes Paper."

If the quadratic approximation to the rate of reaction term suggested by Gray and Harper is applied to Equation 2 and if the subsequent equation is made dimensionless, the result is Equation 10A.

$$\frac{d^2\phi}{dy^2} = -\delta [1 + (e - 2)\phi + \phi^2] \quad (10A)$$

At attempt to solve Equation 10A resulted in an integral which could not be evaluated. After checking integral tables thoroughly, it was concluded that Equation 10A is not solvable in closed form.

STEADY-STATE TEMPERATURE GRADIENTS

The steady-state temperature profiles were determined for a 1-inch-thick infinite slab of RDX at a number of hot-plate temperatures between 400°K and 433°K . The physical parameters listed by Zinn and Mader were used. Equation 9 was used for these computations. The minimum temperature of explosion, that is to say, the maximum hot-plate temperature which will permit a steady state to exist for the system described above is 433°K .

Calculations for a hot-plate temperature of 400°K revealed a temperature rise at the center of 0.036°K above the hot plate, a negligible "self-heating effect."

An attempt was made to find a solution at 435°K , which is above the minimum temperature of explosion. Figure 4 illustrates the fact that there is no solution for the center temperature. The point of closest approach seems to be at about 445°K . On either side of this temperature, the curves for the "e" term and the " \tanh^{-1} " term diverge.

Solutions were obtained for hot-plate temperatures of 410° , 420° , 425° , and 430°K . The graphical solution for the center temperature when the hot-plate temperature is 420°K is illustrated in Figure 5. This type of graphical solution merely avoids trial-and-error procedures for ϕ_0 . Figure 6 shows the temperature profile for hot-plate temperatures of 410°K and 420°K , and Figure 7 gives the same information for 425°K and 430°K . These graphs illustrate several facts which were apparent from the statement of the problem and the solution, namely, that the temperature distribution is symmetrical about the center plane and that the maximum temperature occurs at the center plane. It can be seen from Figure 7 that, for a hot-plate temperature of 430°K , the temperature distribution is quite well described by a parabola. A parabola was forced through the points $T = 433.4$, $x = 0$ and $T = 430.0$, $x = 1.25$ and the resulting points are shown on the figure.

Calculation of the minimum temperature of explosion ($T_{HP} = 433^{\circ}\text{K}$) for this case showed that the corresponding center temperature was 442.25°K or 9.25°K above the hot-plate temperature. This temperature rise is the maximum that can be obtained at a steady state for the parameters studied here. Thus, one can conclude that the

approximation for the heat-generation term, which was used to facilitate solution of the differential equation, is satisfactory for these particular conditions. It was calculated, using $E = 47,500$ cal/g mole, that the approximation $e^{-E/RT_{HP}} \approx e^{E(T - T_{HP})/RT_{HP}^2}$ describes the "true" rate $e^{-E/RT}$ to within 2.5% for this maximum temperature rise of 9.25°K and a hot-plate temperature of 433°K . One would estimate from Figure 3 that the approximation in this case would be within about 3% in spite of the fact that Figure 3 was made for an activation energy of 65,000 cal/g mole rather than the 47,500 of interest here and for hot-plate temperatures of around 600°K rather than of 433°K .

Figure 8 summarizes the maximum center temperatures as the temperature of the hot plate is increased. The rise begins from almost nothing at 400°K to 9.25°K at the minimum temperature of explosion of 433°K . If the hot-plate temperature were increased beyond 433°K , a steady state could not be supported, the center temperature would tend toward infinity, and an explosion would result.

CRITICAL PARAMETERS

Most authors who have considered the steady-state problem have concentrated on the critical parameters -- those values of the environment and the geometry of the explosive itself which just barely permit a steady state to exist. These efforts have been concentrated on obtaining critical values for the dimensionless rate parameter, δ . The critical value, δ_c , is the largest value of δ for which a steady state can exist. A solution for the minimum temperature of explosion which is not explicit in T_m , involving the parameter δ_c , is shown in Equation 11.

$$T_m = \frac{E}{2.303 R \log \frac{\rho Q Z a^2 E}{\lambda R T_m^2 \delta_c}} \quad (11)$$

To solve Equation 11 for T_m , one needs a number for δ_c . The value for δ_c depends on the geometry of the explosive and its environment. Most of the published papers give values for δ_c .

If the solution for the center temperature in Equation 9 is considered with y equal to 0, whence ϕ becomes ϕ_0 , the result is

$$-1 = \sqrt{\frac{2e^{-\phi_0}}{\delta}} \left[\tanh^{-1} \sqrt{1 - e^{-\phi_0}} \right] \quad (11A)$$

If this equation is considered in terms of the physical parameters rather than the dimensionless variables, it can be seen that for a given explosive, the physical and chemical properties E , ρ , Q , Z , and λ are constant. Thus, when a particular explosive is studied, only the center temperature, T_0 , the hot-plate temperature, T_{HP} , and the half thickness, "a", are possible variables. For a given hot-plate temperature, T_0 will then be a function of only the half thickness.

A hypothetical experiment could be considered in which a large hot plate has on its surface a number of slab samples of explosive each thicker than the last. One could observe which ones explode, that is to say, which ones cannot support a steady state, and, in the ones which can support a steady state, one could measure the center temperature. It should be expected that the thickest ones will explode and that the thinnest one will have the least rise above the hot-plate temperature. The results of calculations of the sort described for

the hypothetical experiment are shown in Figures 9 and 10 for infinite slabs exposed to a constant hot-plate temperature of 430°K . Figure 9 shows that, for a half thickness of 1.25 cm, the center temperature is 433.4°K , as shown in Figure 7 as well. However, if the explosive half thickness had been 1.52 cm, a 25% increase, it would have exploded at a hot-plate temperature of 430°K . As the slab becomes thicker, it becomes more and more difficult for the heat to escape; hence, it builds up to explosive proportions. As the slab thickness approaches zero, the center becomes nearer to the hot plate and is, therefore, more easily cooled by the hot plate. At zero thickness, the center temperature must be that of the hot plate, as it is. Figure 10 is shown to complement Figure 9 merely because the half thickness is given in the expression for δ as the square rather than the first power.

The maximum or minimum conditions for Equation 11A could thus be determined by getting the derivative $\frac{da^2}{dT_0}$ and setting it equal to zero. Considering the definitions of the dimensionless variables it can be shown that setting $\frac{da^2}{dT_0} = 0$ is equivalent to

setting $\frac{d\delta}{d\phi_0} = 0$, which is procedurally more attractive.

This procedure results in a single transcendental equation with one unknown, ϕ_0 . The solution from this work for ϕ_0 is shown in Figure 11. The result is $\phi_0 = 1.187$. When this result is substituted into Equation 11A, it is found that the critical δ is 0.8784.

A table of the critical values resulting from the steady-state solution to Equation 8, with boundary conditions 3 and 4 in dimensionless form, follows:

TABLE 2

Critical Parameters for the Steady State

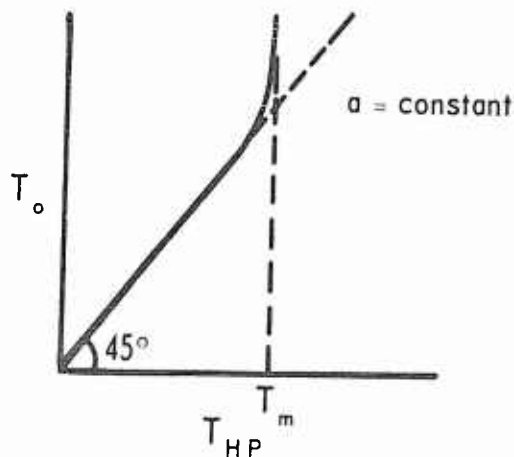
<u>Geometry</u>	<u>ϕ_0</u>	<u>δ_c</u>	<u>Investigator</u>
Slab	1.187	0.8784	Stein
Slab	-----	0.88	Frank-Kamenetsky
Cylinder	-----	2.00	Frank-Kamenetsky
Sphere	-----	3.32	Frank-Kamenetsky
Cylinder	1.39	2 (Integer)	Chambre
Sphere	1.61	3.32	Chambre
Slab	-----	0.87846	Enig et al

The neat condition of merely having a single number for the value of δ is not realized in two slightly more complicated cases: (a) the exact description of the rate equation by $e^{-E/RT}$ and (b) boundary conditions other than 3 and 4 such as 5 and 6 or even the symmetrical heating case of 4 and 6.

Enig, Shanks, and Southworth in Second Ignition Symposium, p. 145, used numerical integration to solve the problem without using an approximation for the rate term. This technique yields a series of numbers for δ_c . δ_c is then a function of the temperature difference between the hot plate and the center. A solution for the minimum temperature of explosion then becomes a slight trial-and-error problem. At low hot-plate-to-center temperature differences, the values of δ_c are, of course, close to those of Chambre.

Thomas and co-workers (Trans Far Soc 57, 2007, 1961; 54, 60, 1958) handled the problems of (1) nonsymmetrical temperature profiles and (2) a symmetrical temperature profile with a temperature discontinuity between the surface and the bulk environment controlled by a surface heat-transfer coefficient. Both of these cases lead to values of δ_c which are a function of the ratio ha/λ . Graphs of these functions make working with them appear fairly easy.

The case as stated earlier in this report has been concerned with considerations of the variation of T_o with half thickness at a constant hot-plate temperature. It is reasonable to ask whether the same relationships will hold if "a" is held constant and the variation of T_o with T_{HP} is studied. At a constant "a", one would expect the following qualitative picture: a nearly straight 45-degree line between T_o and T_{HP} at low values of T_{HP} with a



sudden increase in T_o culminating in explosion. Hence, to find the critical conditions for explosion in this case one would hold "a" constant, get the derivation dT_{HP}/dT_o and set it equal to zero. Fortunately, this procedure yields exactly the equation obtained by considering the variation of T_o with "a" at constant T_{HP} .

EFFECTS OF VARIABLES ON MINIMUM TEMPERATURE OF EXPLOSION

Equation 11 for the minimum temperature of explosion was investigated for the effects of individual variables. It is to be emphasized here that Equation 11 is not proprietary to any particular author. If one solves Equation 8 for $k = 0$, with boundary conditions 3 and 4, and then applies the condition that a solution must exist, the analytical result is Equation 11.

The results of the study are shown in Figures 12 and 13 where the activation energy is held constant, and in Figure 14, where all variables but the activation energy are held constant. Figure 13 differs from Figure 12 only in that E is contained in the ordinate of 13 and is not used at all in 12. It is easily seen that increases in ρ , a , Q , and Z lead to decreases in the minimum temperature of explosion, while increases in λ , R , and δ_c (progression from slab (0.88) to spherical (3.32) geometry) lead to increases in the minimum temperature of explosion. The effect of E is shown in Figure 14. As E increases, T_m increases, if all other parameters are constant.

It is interesting to study the effect of variables over all ranges from zero to infinity and not just in the range which currently seems to be reasonable. The following paradox makes this study of interest. If an explosive has zero thermal conductivity, then, when it is suddenly exposed to the hot plate, it will retain its initial temperature because no heat can be transferred into the explosive. Hence, no explosion can occur no matter what the hot-plate temperature. The minimum temperature of explosion is thus infinite. However, Figures 12 and 13 show that a decrease in the thermal conductivity leads to a decrease in the minimum temperature of explosion. This paradox is neatly resolved by studying the entire range of thermal conductivities from zero to infinity.

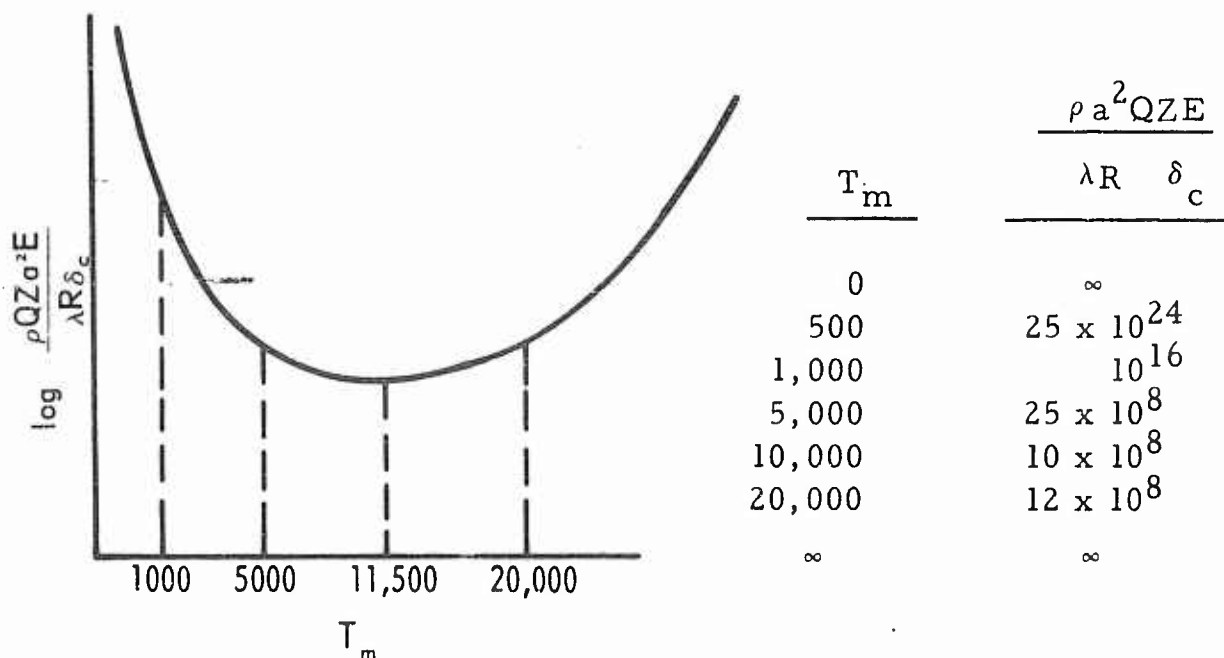
If E is held constant and the variables Q , Z , ρ , a^2 , $\frac{1}{\lambda}$, and $\frac{1}{\delta_c}$ in Equation 11 are differentiated with respect to T_m , one at a time while all other values are held constant, it is found that a minimum value for T_m is reached at $T_m = E/2R$. E is often a number like 46,000 cal/g mole, hence, a typical minimum in the curve of log

$$\frac{\rho a^2 Q E Z}{\lambda R \delta_c} \quad \text{versus } T_m \text{ occurs at } 11,500^\circ\text{K, a minimum}$$

temperature of explosion which is out of the question. Further investigation of Equation 11 reveals that, as λ approaches zero or ρ , a , Q , and Z become very large (i.e., the term

$\frac{\rho a^2 QZ}{\lambda R \delta_c}$ approaches infinity), then the equation can be satisfied if T_m approaches zero. On the other hand, Equation 11 is also satisfied if the terms $\frac{\rho a^2 QZ}{\lambda R \delta_c}$ and T_m^2 both approach

infinity at the same rate, so that the argument of the logarithm approaches unity. These observations plus the fact that a minimum exists in the curve at $E/2R$ led to the following qualitative figure:

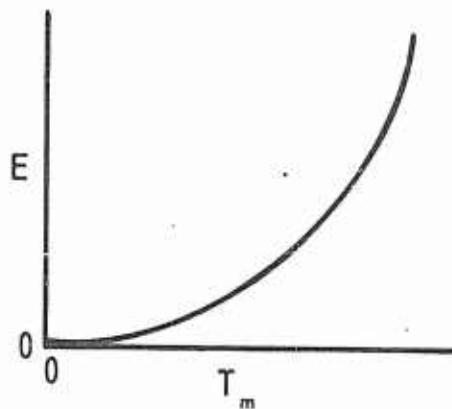


If one takes $E = 46,000$ cal/g mole, the table to the right of the figure is obtained. It may be observed, for what it is worth, that the minimum is very shallow and is quite flat between say $5,000^\circ\text{K}$ and $20,000^\circ\text{K}$. The figure further emphasizes that Equation 11 is double valued in T_m . This point may be important in computer work. The computer may be very happy with a ridiculous, although entirely correct, solution to Equation 11 like $50,000^\circ\text{K}$ when the other root of, say 473° , would be the meaningful one.

It seems logical to ask, in conjunction with the RDX calculations, how small "a," the half thickness of an infinite slab must be in order to reach the minimum in the curve if all other variables have the values given by Zinn and Mader. The answer is approximately 10^{-10} cm.

Similarly, one might ask at what value a decrease in the thermal conductivity begins to cause a rise in the minimum temperature of explosion toward its value of infinity when $\lambda = 0$. For the one-inch infinite slab of RDX with all other variables having the values assigned by Zinn and Mader, the thermal conductivity must be approximately 10^{-23} cal/cm sec^oK and decreasing in order to cause an increase in the minimum temperature of explosion.

Investigation of Equation 11 was also carried out for all ranges of activation energy with all other variables held constant. There is no relative maximum or minimum in E with respect to T_m . The following qualitative figure summarizes the results:



With regard to this figure, the following observations may be made: (1) At $E = 0$, $T_m = 0$ and the slope dE/dT_m equals 0, and (2) as E approaches ∞ , T_m approaches ∞ , and the slope dE/dT_m approaches ∞ .

REVIEW OF THOMAS AND BOWES PAPER

The paper by Thomas and Bowes in Trans Far Soc 57, 2007, 1961 has been reviewed in considerable detail. This case is closer to the reality of the experiment conducted by the Explosives Research Section at Picatinny than any other currently available. These authors solved Equation 8 for the infinite slab with boundary conditions 5 and 6 in dimensionless form. Equation 8, of course, includes the exponential approximation for the Arrhenius term. In the majority of the cases, the Thomas and Bowes equations were neater than those obtained by this review, and in all cases they certainly exhibited more polish. However, a temperature profile for subcritical conditions can be obtained much more easily from the equations which follow in this report than from those of Thomas and Bowes. Equation 12 is the solution to the differential equation.

$$y = \sqrt{\frac{2}{\beta}} \left[\tanh^{-1} \sqrt{1 - \frac{\delta}{\beta}} - \tanh^{-1} \sqrt{1 - \frac{\delta e^{\phi}}{\beta}} \right] \quad (12)$$

where

$$\beta = \delta e^{\phi_{2a}} + \frac{a^2}{2} (\phi_{2a} - \phi_{\infty})^2.$$

Equation 13 results from Equation 12 if only the maximum value of ϕ_m is at issue.

$$e^{\phi_m} = e^{\phi_{2a}} + \frac{a^2}{2\delta} (\phi_{2a} - \phi_{\infty})^2 \quad (13)$$

In Equation 12, a , ϕ_{∞} , and δ (less than δ_c) are constants. To get a temperature profile, one sets $y = 2$, which makes ϕ become ϕ_{2a} in Equation 12. ϕ_{2a} is the only remaining unknown, and so its value is obtained. Substitution of a number for ϕ_{2a} into Equation 12 leaves only the unknowns y and ϕ . Hence, insertion of various values of ϕ gives the $y - \phi$ curve (i.e., the temperature profile). If the maximum temperature and the position of the maximum temperature are of interest to the exclusion of the remainder of the profile, one can proceed by inserting the number for ϕ_{2a} into Equation 13 and evaluating ϕ_m directly. This value of ϕ_m along with ϕ_{2a} gives y_m directly from Equation 12.

The use of the Thomas and Bowes solution, their Equations 5, 6, and 8i, requires simultaneous solution of two transcendental equations, their Equations 6 and 8i, for the maximum values, ϕ_m and \bar{Z}_m . These numbers can be inserted into their Equation 5 in order to get the temperature profile, $\phi - \bar{Z}$ curve. Hand solution of two transcendental equations, simultaneously, for two variables seems to be quite a formidable task.

Thomas and Bowes were primarily interested in obtaining critical parameters for the problem they were investigating. The critical parameter δ_c is a function of two variables, a and the environmental temperature. As a result, their solution is approximate but very close for values of a greater than 0.5, which includes all realistic values of a . A graph of their solution appears in the paper.

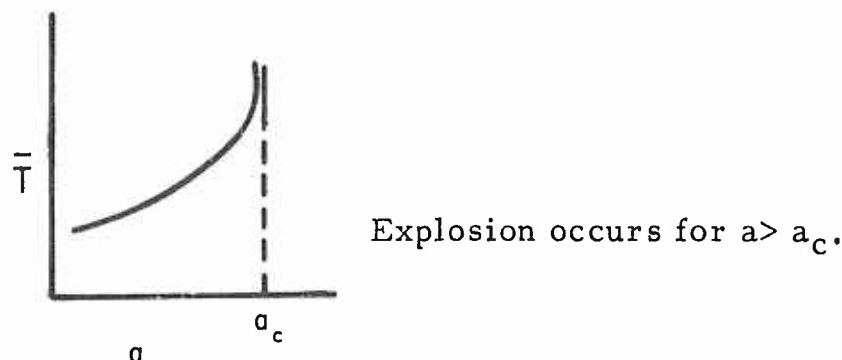
Considerable effort was spent looking into the problem of maxima and minima of a function of two variables in order to try to arrive at an exact solution for all positive values of a . After considerable work, it was concluded that this particular problem has no exact solution for all values of a . Furthermore, it became apparent that no single maximum δ exists for all values of a and ϕ_∞ even if the problem could be solved rigorously. It is believed as Thomas and Bowes graph indicates, that δ increases continuously with a as a approaches infinity.

REVIEW OF SEMENOV SOLUTION OF STEADY-STATE PROBLEMS

One of the first investigations of the steady-state problem was made by Semenov (actually the Semenov paper was not used; Thomas, Trans Faraday Society 56, 833, 1960 was used as a guide to Semenov's work), who studied it in terms of average temperatures in the explosive; that is to say, the temperature was assumed not to vary with position within the explosive. If one assumes the temperature to be everywhere the same within an infinite slab of explosive and that all heat generated within the explosive is transferred to the surroundings, Equation 14 results.

$$a \rho Q Z e^{-E/R\bar{T}} = h (\bar{T} - T_{\infty}) \quad (14)$$

This equation is also the solution for \bar{T} . For a constant ambient temperature, the temperature of the explosive increases with increasing slab thickness until a certain thickness is reached at which this slab will explode. Graphically, this relationship is:



To obtain the critical temperature rise one needs to obtain $da/d\bar{T}$ from Equation 14 and set it equal to zero. This procedure results in a critical temperature rise of $\phi = 1$ or, expressing ϕ in its defining terms,

$$(\bar{T} - T_{\infty}) = \frac{RT_{\infty}^2}{E} \quad (15)$$

It is not necessary to apply any approximation to the rate term in Equation 14 in order to perform the required mathematical operations. However, the exponential approximation has been used to be consistent with previous calculations. Without this approximation, the results would be the same except that \bar{T} would replace T_{∞} on the right-hand side of the above equation. The solution for the critical half thickness then becomes

$$\frac{h}{a_c e} = \left(\frac{E}{RT_\infty} \right)^2 Q \rho Z e^{-E/RT} \quad (15A)$$

Manipulation of this equation into the form of the one for the minimum temperature of explosion in the spatial-varying temperature problem is given in Equation 16.

$$T_{\infty m} = \frac{E}{2.303 R \log \frac{\rho Q Z a E e}{h R T_{\infty m}}} \quad (16)$$

Equation 16 should be compared to Equation 11, which is the spatial-varying-temperature solution. The surface heat-transfer coefficient replaces the thermal conductivity; the half thickness appears only to the first power; and the geometric term δ is absent. The constant e also has been introduced.

Further manipulation of Equation 15A to introduce the dimensionless rate parameter δ shows the critical value of for the infinite slab to be $\delta_c = \frac{a}{e}$.

SYMBOLS

ρ = density of explosive, g/cm³

Q = heat of reaction, cal/g

Z = frequency factor, sec⁻¹

E = activation energy, cal/g mole

R = universal gas constant, cal/g mole °K

T = absolute temperature, °K

λ = thermal conductivity, cal/cm sec °K

∇^2 = Laplacian operator

C = specific heat of explosive, cal/g °K

t = time, sec

a = half thickness of explosive or radius of cylinder or sphere, cm

T_{HP} = absolute temperature of hot plate, °K

x = distance into explosive, cm

h = heat transfer coefficient, cal/cm² °K sec

T_{2a} = absolute temperature at distance $2a$, °K

T_{∞} = absolute temperature of surroundings, °K

ϕ = dimensionless temperature = $\frac{E}{R T_{HP}^2} (T - T_{HP})$

y = dimensionless distance = $\frac{x}{a}$

δ = dimensionless reaction rate = $\frac{a^2 E \rho Q Z e^{-E/RT_{HP}}}{R T_{HP}^2 \lambda}$

ϕ_0 = ϕ at position $x = 0$

$$\bar{z} = \text{dimensionless distance} = \frac{r}{a}$$

$$T_m = \text{minimum (hot plate) temperature of explosion, } ^\circ\text{K}$$

$$T_o = \text{temperature at center of slab, cylinder, or sphere, } ^\circ\text{K}$$

$$\phi_{2a} = \phi \quad \text{at position } x = 2a$$

$$a = \text{dimensionless heat-transfer ratio} = ha/\lambda$$

$$\phi_\infty = \phi \quad \text{at the bulk temperature of the environment}$$

$$\phi_m = \text{maximum value of } \phi$$

$$y_m = \text{dimensionless position at which } \phi_m \text{ occurs}$$

$$\bar{T} = \text{average absolute temperature in explosive, } ^\circ\text{K}$$

$$T_\infty = \text{absolute temperature of surroundings, } ^\circ\text{K}$$

$$T_{\infty m} = \text{minimum temperature of explosion for Semenov solution, } ^\circ\text{K}$$

$$\tanh^{-1} = \text{inverse hyperbolic tangent}$$

$$\delta_c = \text{critical value of } \delta$$

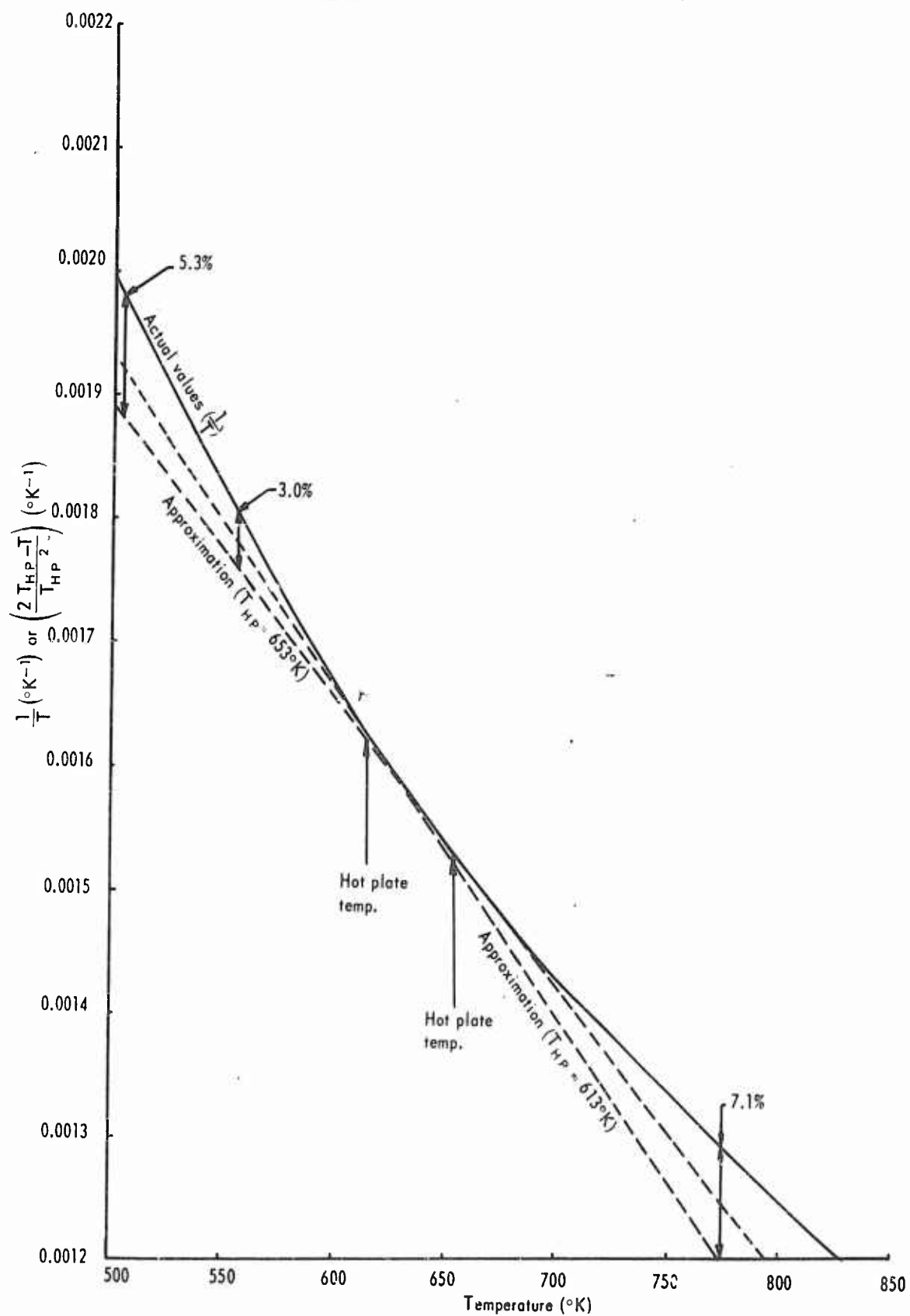


Fig 1 Comparison of $\frac{1}{T}$ to its approximation $\frac{2T_{HP} - T}{T_{HP}^2}$

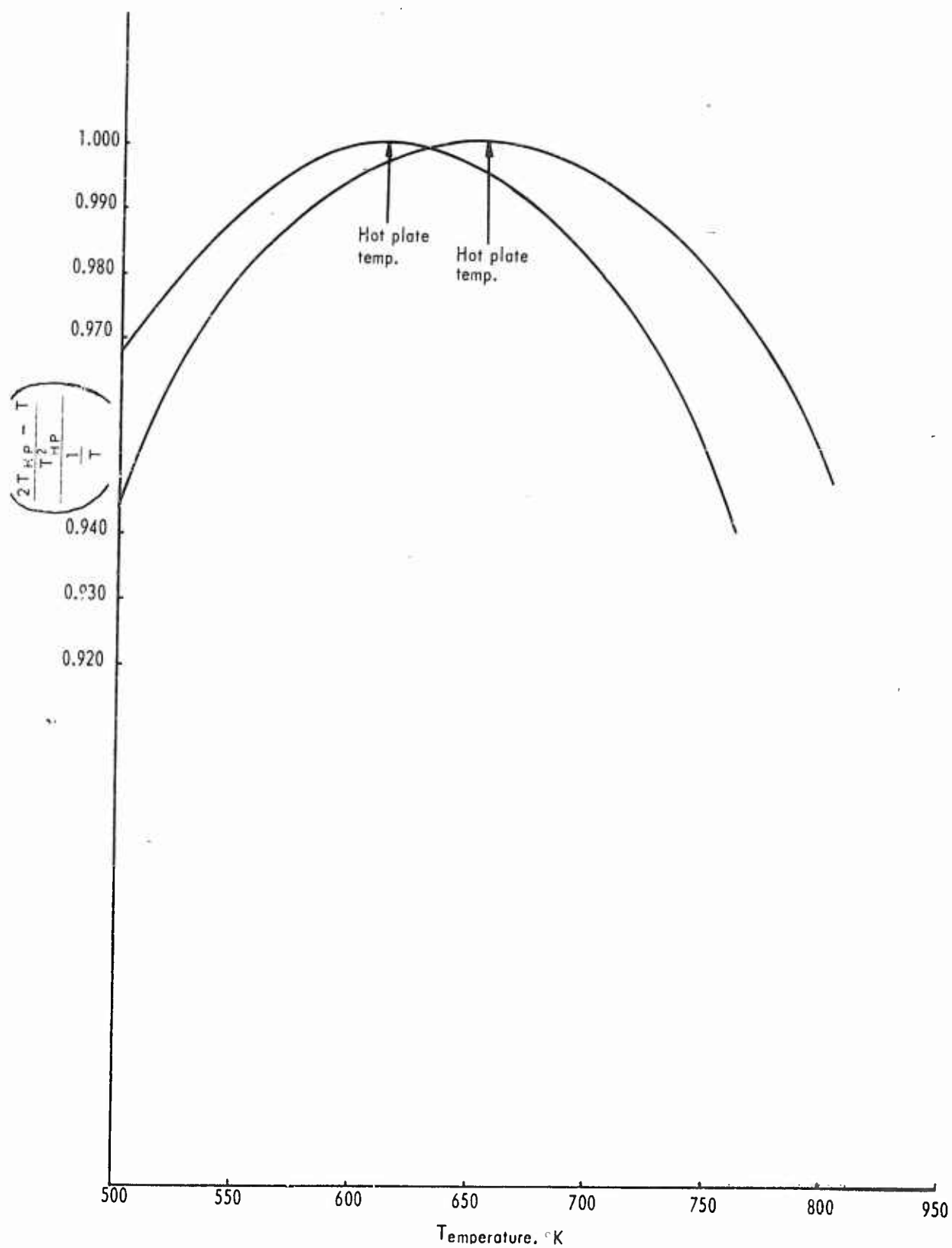


Fig 2 Comparison of $\frac{1}{T}$ to the ratio of its approximation $\frac{2 T_{HP} - T}{T_{HP}^2}$ to $\frac{1}{T}$

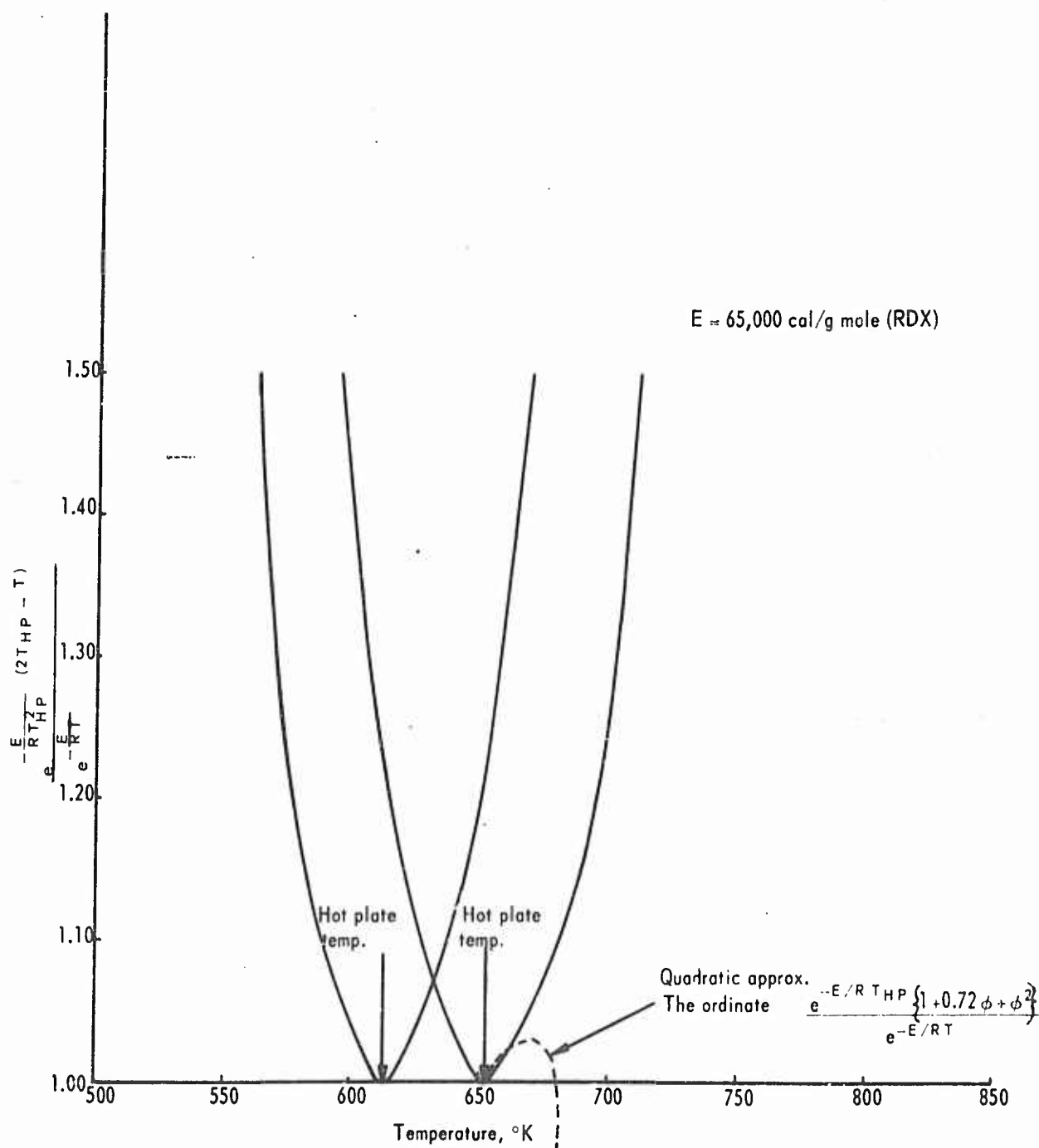


Fig 3 Comparison of $e^{-\frac{E}{RT}}$ to its approximation $e^{-\frac{E}{RT_{HP}}(2T_{HP} - T)}$

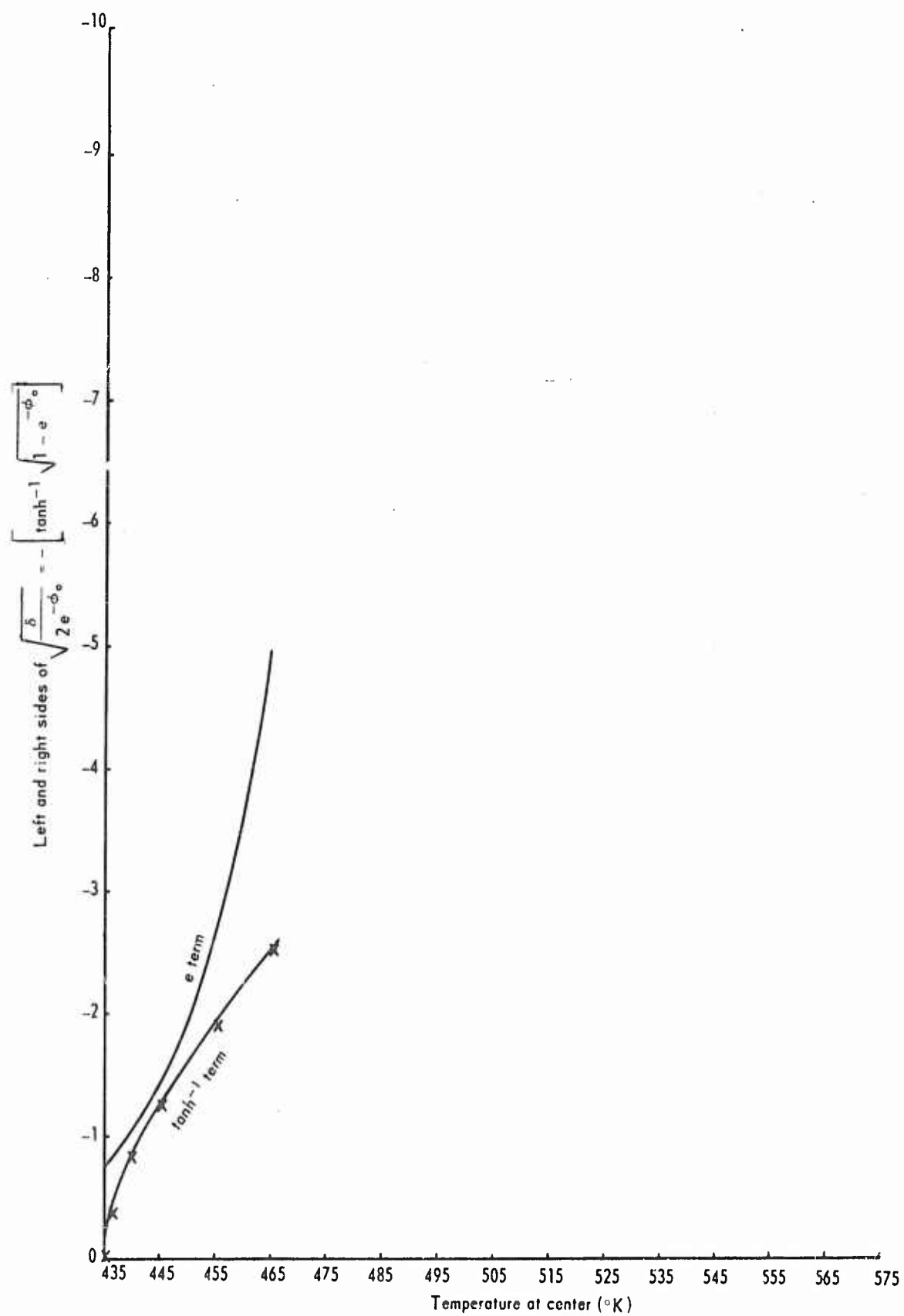


Fig 4 Attempt to find solution to Equation 9 for $T_{HP} = 435^{\circ}\text{K}$, a temperature for which no solution exists

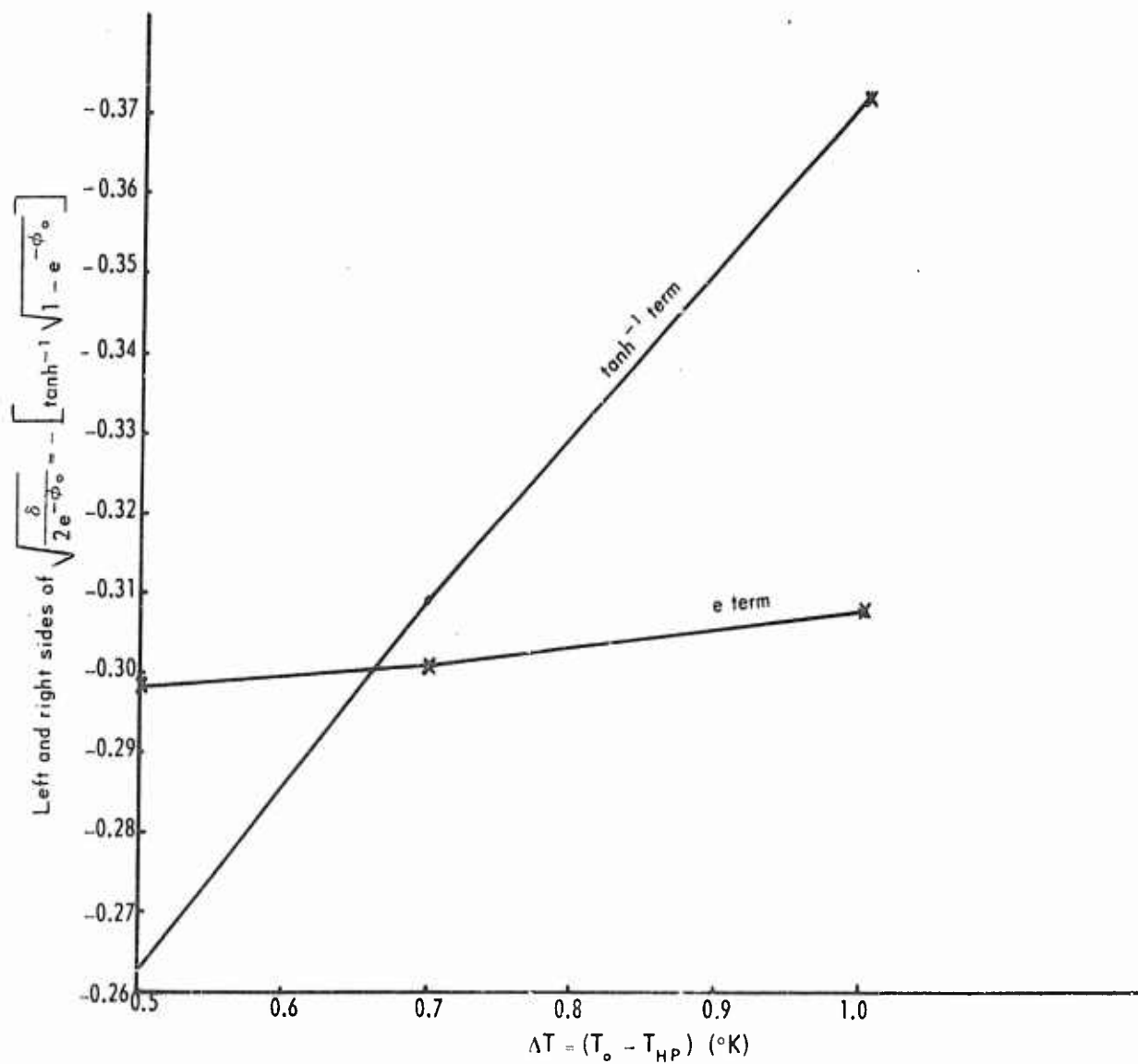


Fig 5 Solution to Equation 9 for $T_{HP} = 420^{\circ}K$

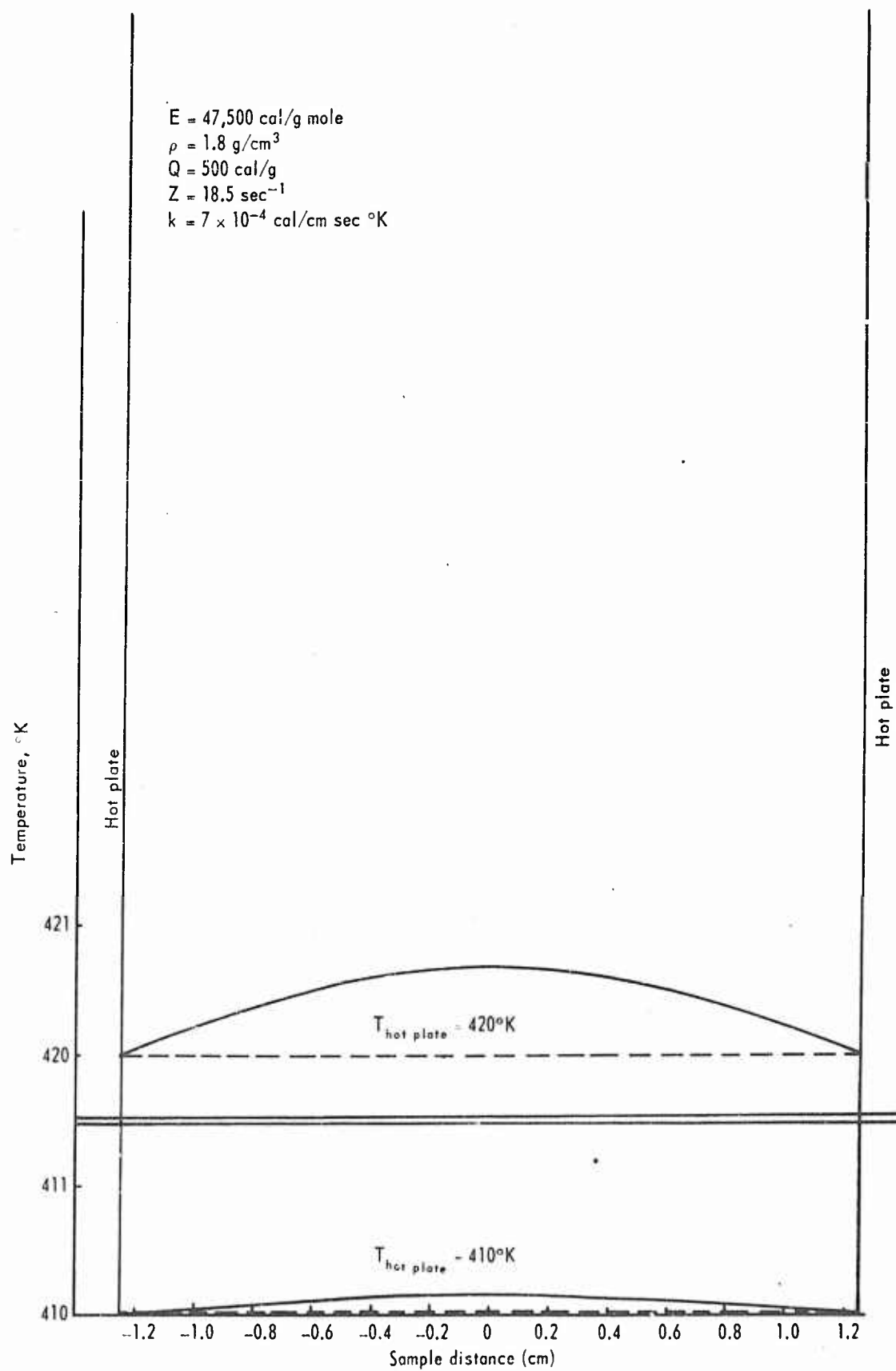


Fig 6 Steady state temperature profiles in 2.50-cm-thick RDX slab

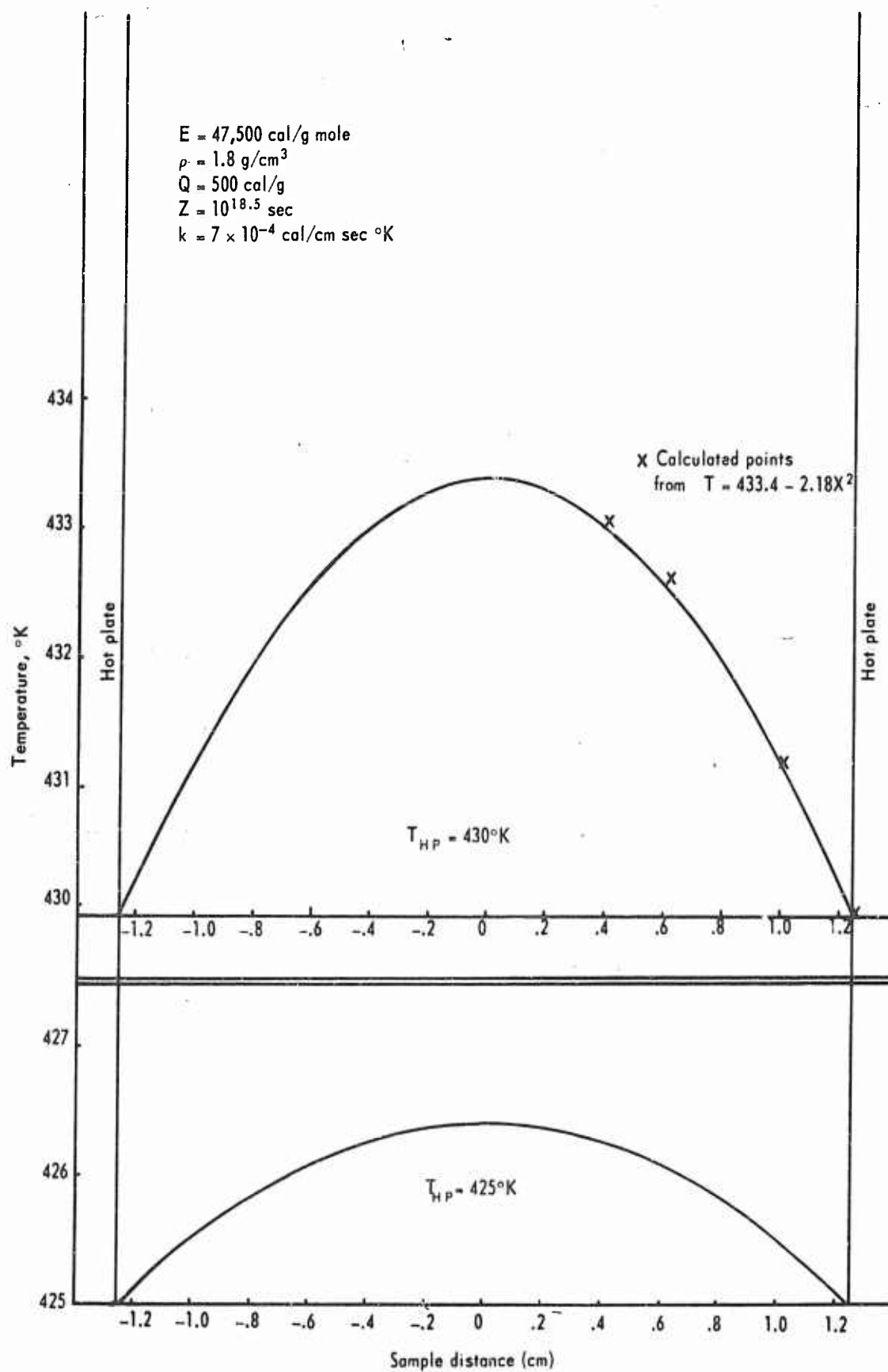


Fig 7 Steady state temperature profiles in 2.50-cm-thick RDX slab

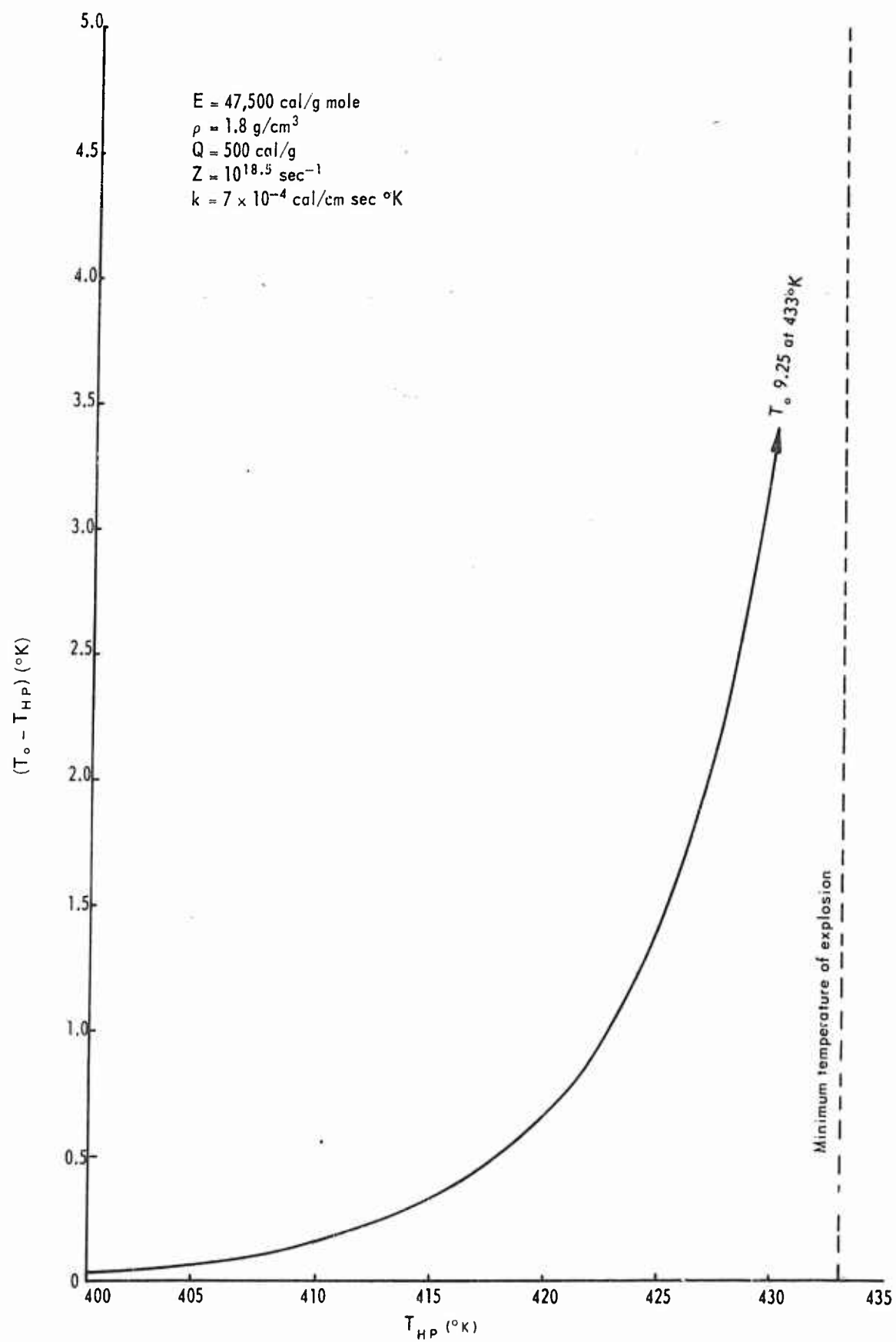


Fig 8 Steady state temperature at center of 2.50-cm-thick RDX slab as a function of hot-plate temperature

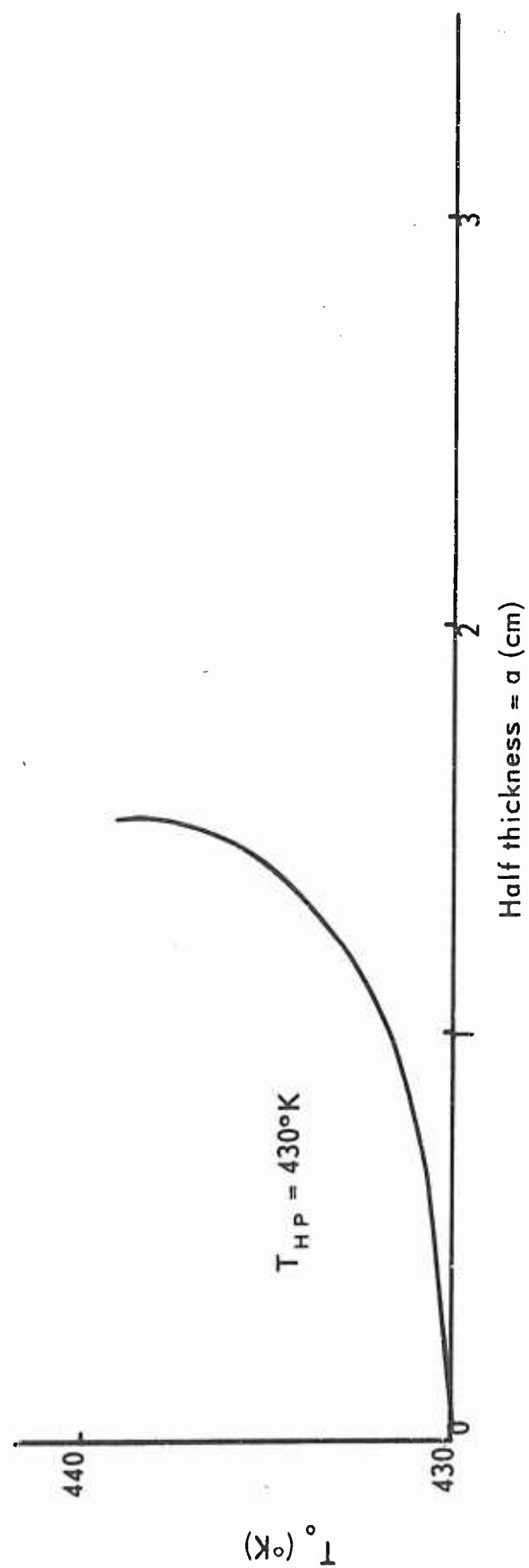


Fig 9 Steady state temperature at center of various thicknesses of RDX slabs for a constant hot-plate temperature of 430°K

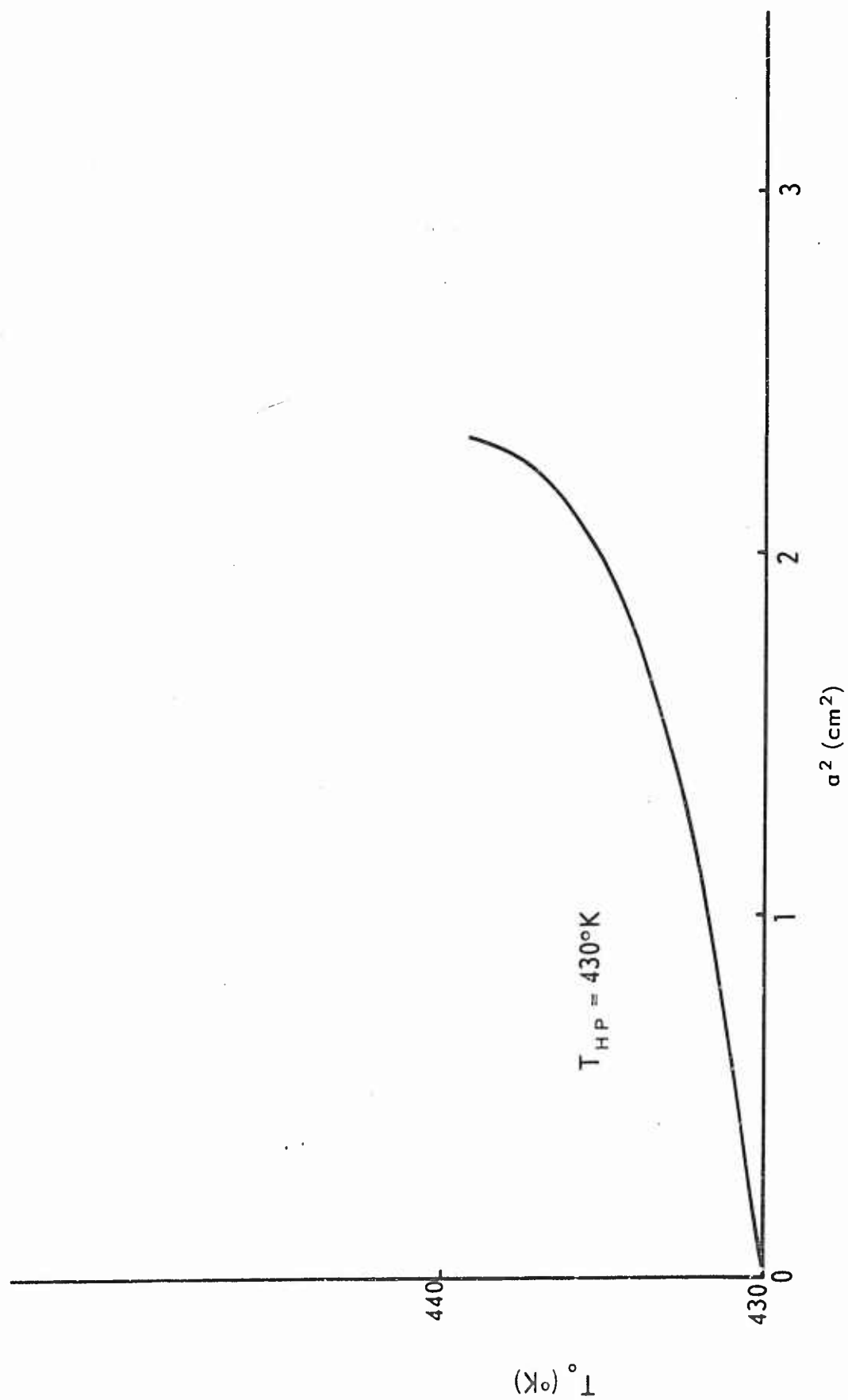


Fig 10 Steady state temperature at center of various thicknesses of RDX slabs for a constant hot-plate temperature of 430°K

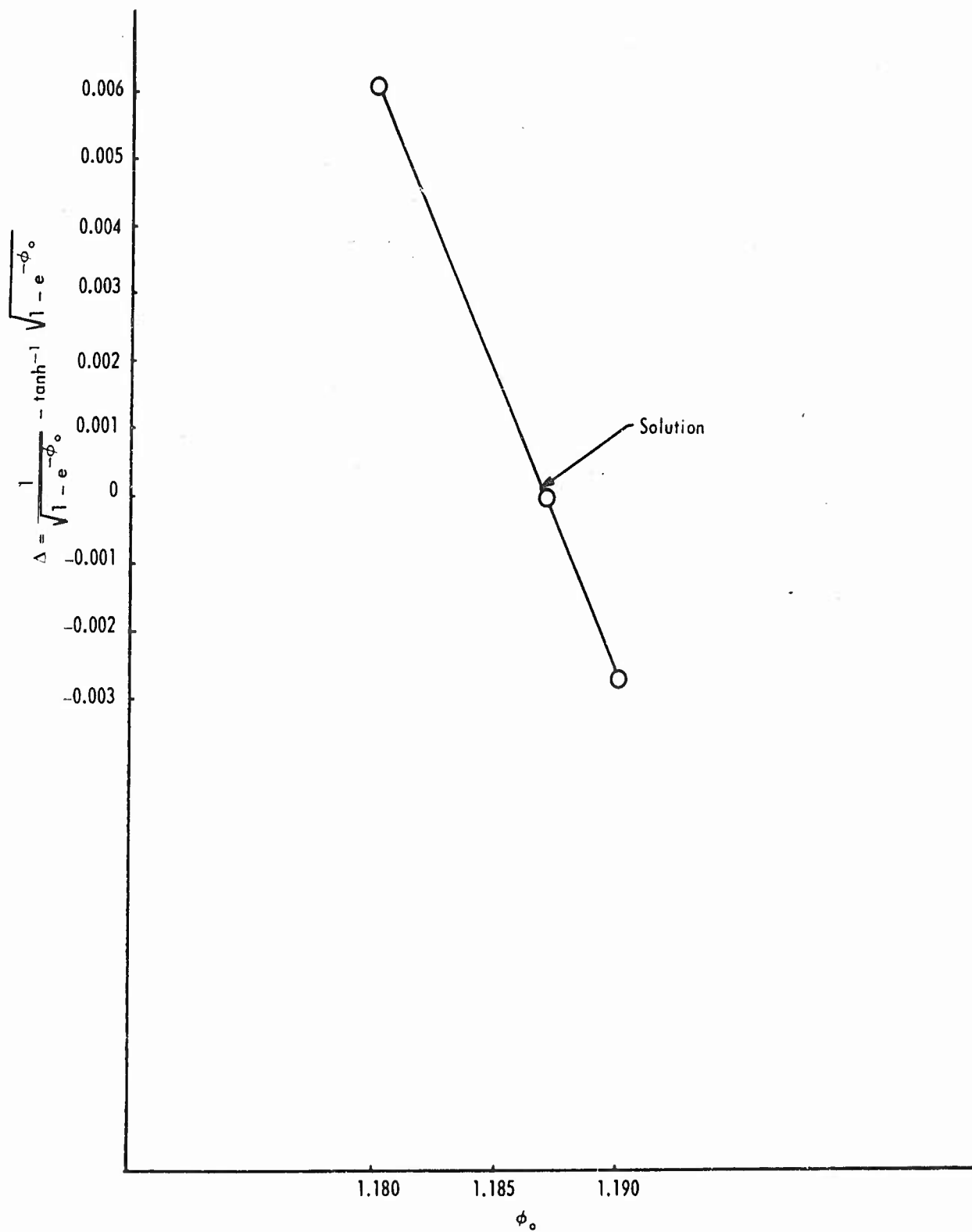


Fig 11 Solution for $\frac{1}{\sqrt{1-e^{-\phi_0}}} = \tanh^{-1} \sqrt{1-e^{-\phi_0}}$

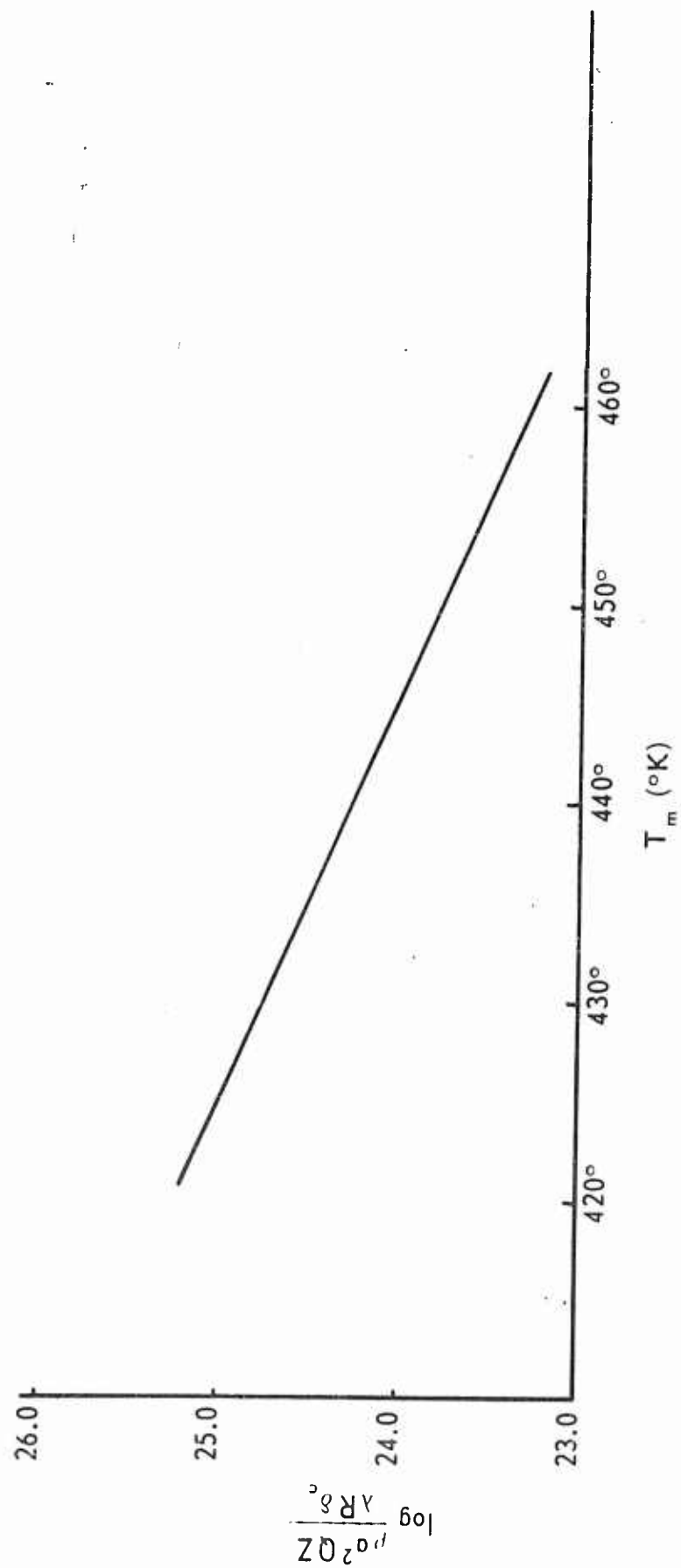


Fig 12 RDX cylinder geometry with E held constant at 47,500 cal/M as a function of $\log \frac{\rho a^2 Q Z}{\lambda R \delta_c}$

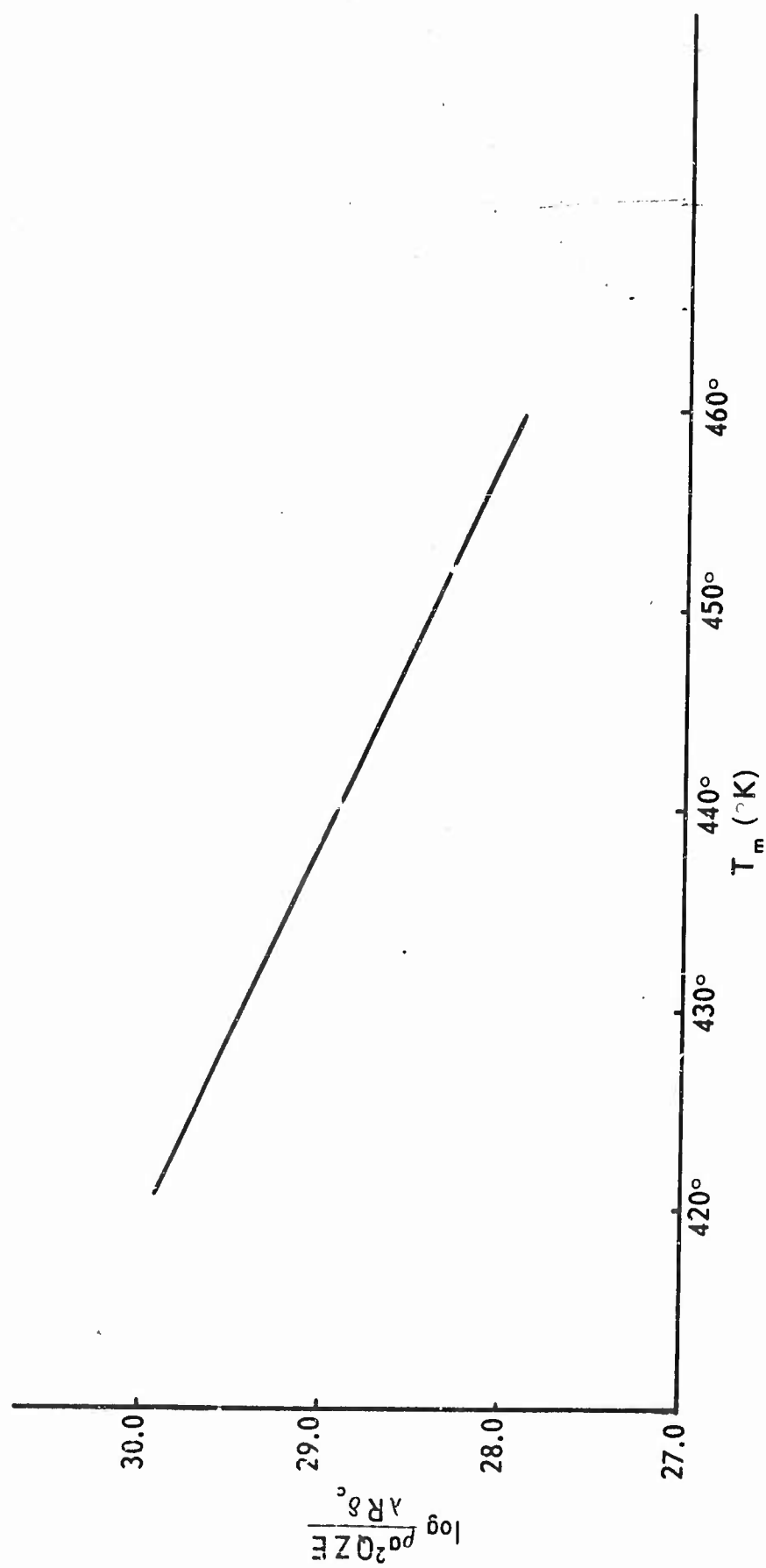


Fig 13 RDX cylinder geometry with E held constant at 47,500 cal/M as a function of $\log \frac{\rho^2 QZE}{\lambda R \delta_c}$

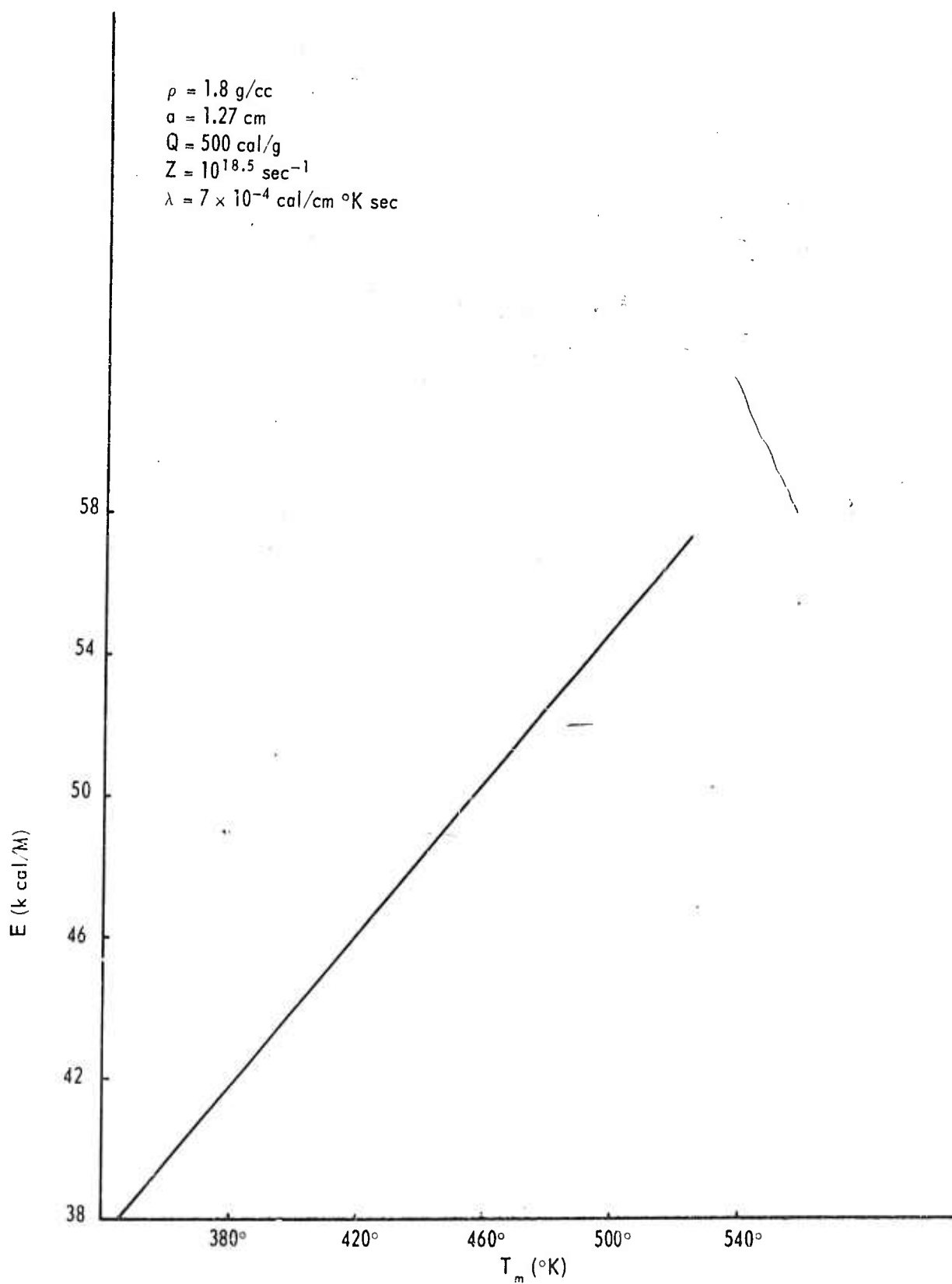


Fig 14 RDX slab geometry with ρ , a , Q , Z , λ held constant

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EXTERNALLY TO ELEVATED TEMPERATURES

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13. ABSTRACT

✓ A critical review of the literature on the minimum temperature for explosion is made and the more important contributions are discussed. The relative importance of the various parameters involved is given, including explosive properties and sample environment, configuration, and bulk mass. It is found that the solutions in the literature are adequate and usable although not always lucid. Several of these solutions are expanded upon and discussed in detail. Many graphical representations and examples are given to illustrate the points presented.

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Steady state						
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